Magnetic-field induced superconductor-antiferromagnet transition in lightly doped $R\mathrm{Ba_2Cu_3O_{6+x}}$ (R=Lu, Y) crystals

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The remarkable sensitivity of the c-axis resistivity and magnetoresistance in cuprates to the spin ordering is used to clarify the doping-induced transformation from an antiferromagnetic (AF) insulator to a superconducting (SC) metal in $R\mathrm{Ba_2Cu_3O_{6+x}}$ (R=Lu, Y) single crystals. The established phase diagram demonstrates that the AF and SC regions apparently overlap: the superconductivity in $R\mathrm{Ba_2Cu_3O_{6+x}}$, in contrast to $\mathrm{La_{2-x}Sr_xCuO_4}$, sets in before the long-range AF order is completely destroyed by hole doping. Magnetoresistance measurements of superconducting crystals with low $T_c \leq 15-20$ K give a clear view of the magnetic-field induced superconductivity suppression and recovery of the long-range AF state. What still remains to be understood is whether the AF order actually persists in the SC state or just revives when the superconductivity is suppressed, and, in the former case, whether the antiferromagnetism and superconductivity reside in nanoscopically separated phases or coexist on an atomic scale.

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INTRODUCTION

The transformation, upon charge doping, of an antiferromagnetic (AF) Mott insulator into a superconducting (SC) metal and the role of AF correlations in the appearance of superconductivity have challenged researchers since the discovery of high-T_c superconductivity in cuprates. Is the AF order an indispensable component or a competitor for the high-T_c phenomenon? In a prototype high-T_c cuprate $\mathrm{La_{2-x}Sr_xCuO_4}$, the long-range AF order is destroyed by doped holes way before the superconductivity sets in [1], which has led to a general belief that the spin frustration is a prerequisite for metallic conduction and superconductivity. The destructive impact of static spin order on superconductivity was further supported by the observation of SC suppression at a peculiar 1/8 doping in $\mathrm{La_{2-x}Ba_xCuO_4}$ [2]. On the other hand, spin excitations are often suggested to provide glue for SC pairing, implying the ultimate importance of AF correlations, be they static or dynamic. Besides, the incompatibility of static AF order and SC may be not necessarily a general feature of cuprates. In $R\mathrm{Ba_2Cu_3O_{6+x}}$ (R is a rare-earth element), for instance, the long-range AF order survives up to much higher doping levels than in $\mathrm{La_{2-x}Sr_xCuO_4}$ [3, 4, 5, 6, 7], though the possibility of its coexistence with superconductivity still remains to be clarified.

In strongly anisotropic high-T_c cuprates, the c-axis charge transport appears to be remarkably sensitive to the spin ordering in $\mathrm{CuO_2}$ planes. In $R\mathrm{Ba_2Cu_3O_{6+x}}$ crystals, for example, the c-axis resistivity $\rho_c(T)$ exhibits a steep increase at the Néel temperature T_N [5, 6, 8]. Even relatively weak modifications of the spin structure such as spin-flop or metamagnetic transitions result in surprisingly large changes—by up to an order of magnitude—in the c-axis resistivity of both hole-doped $\mathrm{La_{2-x}Sr_xCuO_4}$ [9, 10] and electron-doped $\mathrm{Pr_{1.3-x}La_{0.7}Ce_xCuO_4}$ [11] and $\mathrm{Nd_{2-x}Ce_xCuO_4}$ crystals [12]. This sensitivity of the interplane charge transport in cuprates to the spin order can be, and actually is, employed for tracing the evolution of the spin state with doping, temperature, or magnetic fields [5, 6, 8, 9, 11].

While electrical resistivity measurements have proved to be a very convenient tool for mapping the magnetic phase diagrams in cuprates, their usage has an obvious limitation; namely, they fail as the superconductivity sets in. Because of this limitation, previous resistivity studies of $R\mathrm{Ba_2Cu_3O_{6+x}}$ crystals [5, 6] could not clarify whether the long-range AF order vanishes by the onset of superconductivity, or extends further, intervening the SC region. It sounds tempting to employ strong magnetic fields to suppress the superconductivity and to use the c-axis resistivity technique of detecting the spin order in otherwise inaccessible regions of the phase diagram. In the present paper, we use this approach to study the very region of the AF-SC transformation in $\mathrm{LuBa_2Cu_3O_{6+x}}$ and $\mathrm{YBa_2Cu_3O_{6+x}}$ single crystals.

EXPERIMENT

$R\mathrm{Ba_2Cu_3O_{6+x}}$ single crystals with nonmagnetic rare-earth elements R=Lu and Y were grown by the flux method and their oxygen stoichiometry was tuned to the required level by high-temperature annealing with subsequent quenching [5, 13]. In order to ensure that no

oxygen-enriched layer was formed at the crystal surface during the quenching process, one of the crystals was dissolved in acid in several steps; resistivity measurements detected no considerable change in the SC transition upon the crystal's surface destruction. The c-axis resistivity $\rho_c(T)$ was measured using the ac four-probe technique. To provide a homogeneous current flow along the c-axis, two current contacts were painted to almost completely cover the opposing ab-faces of the crystal, while two voltage contacts were placed in small windows reserved in the current ones [5]. The magnetoresistance (MR) was measured by sweeping temperature at fixed magnetic fields up to 16.5 T applied along the c axis of the crystals.

RESULTS AND DISCUSSION

A representative $\rho_c(T)$ curve obtained for a $LuBa_2Cu_3O_{6+x}$ single crystal with a doping level slightly lower than required for the onset of superconductivity is shown in Fig. 1. In general, the c-axis resistivity in $R\mathrm{Ba_2Cu_3O_{6+x}}$ crystals of non-SC composition exhibits two peculiar features upon cooling below room temperature, both of which can be seen in Fig. 1. The first one is a pronounced crossover at T_m ($T_m \approx 120\,\mathrm{K}$ for the particular composition in Fig. 1), indicating a change—with decreasing temperature—of the dominating conductivity mechanism from some kind of thermally activated hopping to a coherent transport [5, 6, 8, 13, 14]. It is worth noting that a similar coherent-incoherent crossover was observed in other layered oxides as well [15]. The second feature is a sharp growth of the resistivity associated with the long-range AF ordering [5, 6, 8]. If the crystals were less homogeneous, the low-temperature resistivity upturn would be easy to confuse with a usual disorder-induced charge localization. However, this sharp resistivity anomaly with a characteristic negative peak in the derivative (inset in Fig. 1) is definitely related to the spin ordering at the Néel temperature T_N: it has been traced from the parent compositions $R\mathrm{Ba_2Cu_3O_6}$ with well-known T_N to avoid any doubt in its origin.

In carefully prepared crystals, the AF transitions remain sharp for all compositions, including crystals with very low $T_N \approx 20\,\mathrm{K}$, that is ~ 20 times lower than original $T_{N0} \approx 420\,\mathrm{K}$ in parent $R\mathrm{Ba_2Cu_3O_6}$. It is important to emphasize that the transitions at $T_N \approx 20\,\mathrm{K}$ remain virtually as sharp as in undoped parent crystals even though spin freezing into a spin-glass state is usually expected for such low temperatures and high hole concentrations [16]. Moreover, the impact of the AF ordering on $\rho_c(T)$ does not weaken with decreasing Néel temperature; as can be seen in Fig. 1, the resistivity of the crystal with $T_N \approx 24\,\mathrm{K}$ increases by more than 50% upon cooling below T_N, while in crystals with $T_N > 100\,\mathrm{K}$ the corresponding ρ_c growth does not exceed 15-20% [5, 8].

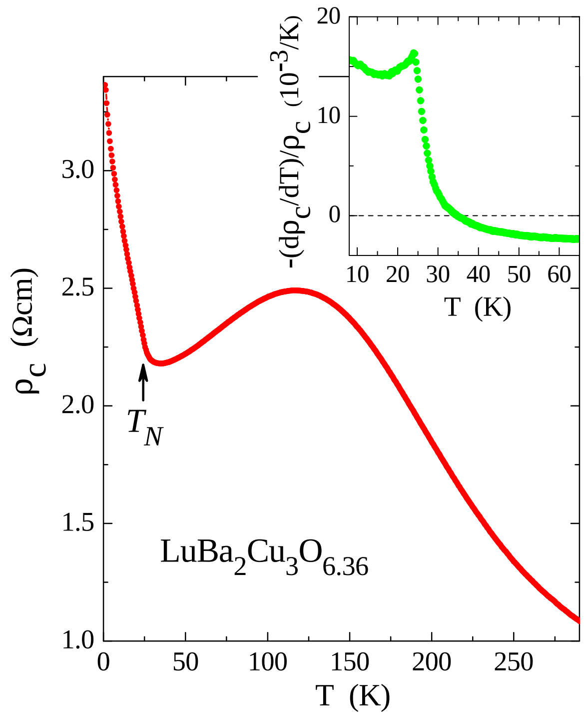

FIG. 1: Out-of-plane resistivity, $\rho_c(T)$, of a $LuBa_2Cu_3O_{6.36}$ single crystal. The sharp growth of the resistivity upon cooling below $\approx 25\,\mathrm{K}$ is caused by the AF ordering. Inset: anomaly in the normalized derivative $(d\rho_c/dT)/\rho_c$ associated with the Néel transition.

What do these observations tell about the impact of doped holes on copper spins? Apparently, the sharp AF transitions are hard to reconcile with strongly frustrated spin states and disordered spin textures in CuO_2 planes that are usually expected to emerge in cuprates with doping [17]. Besides, a strong frustration-induced reduction of the staggered magnetization $M^\dagger$ required to account for the decrease in T_N would necessarily diminish the impact of the interplane spin ordering on ρ_c, which again disagrees with observations. On the other hand, if the role of mobile doped holes is not to introduce a uniform spin frustration, but simply to break the long-range AF order into two-dimensional domains in CuO_2 planes, the observed behavior is easier to understand. In this case, it is the AF domains in CuO_2 planes that become the elementary magnetic units and the long-range AF state should develop through ordering of their phases, which can occur rather abruptly. Correspondingly, the T_N evolution with doping should be governed by the decreasing AF domain size, rather than $M^\dagger$. In turn, the ordering of AF domains whose local staggered magnetization does nor change appreciably with hole doping leaves room for large changes in the c-axis resistivity even at low T_N.

The T_N and T_c values determined from the c-axis resistivity of $R\mathrm{Ba_2Cu_3O_{6+x}}$ crystals have been used to estab-

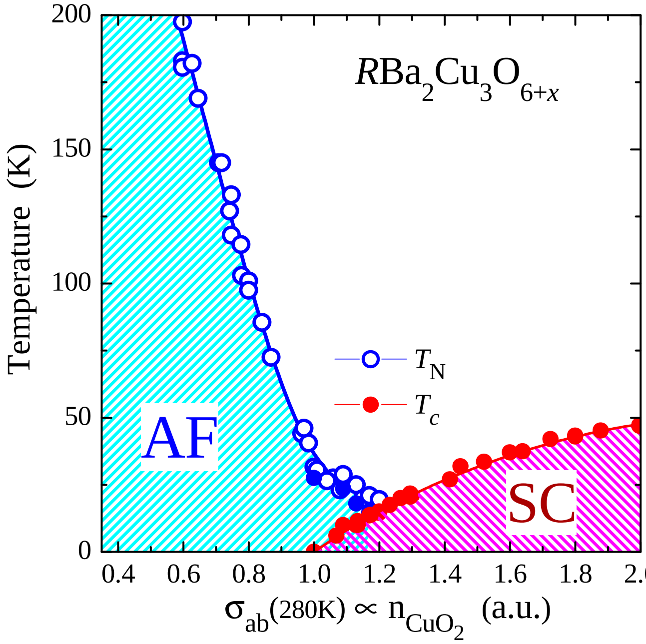

FIG. 2: Phase diagram of $R\mathrm{Ba_2Cu_3O_{6+x}}$ (R = Lu, Tm, Y) crystals near the AF-SC transformation. The AF and SC transition temperatures, T_N and T_c, are presented as a function of the in-plane conductivity $\sigma_{ab}(280K)$ which is a good measure of the hole density in the shown doping region [18]. The Néel temperature was determined either at the position of the jump (middle point) in the derivative $d\rho_c/dT$ (open circles), or at the position of the negative peak in $d\rho_c/dT$ (solid circles).

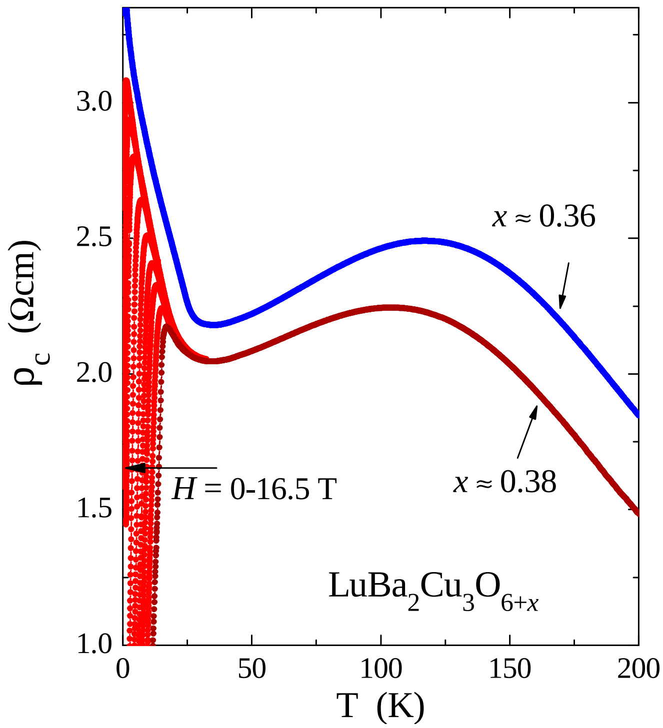

FIG. 3: Out-of-plane resistivity of one and the same $\mathrm{LuBa_2Cu_3O_{6+x}}$ single crystal for two oxygen concentrations near the AF-SC transformation. For the superconducting composition $x = 0.38$, the data were taken at several magnetic fields–from zero up to 16.5 T–applied along the c axis. The sharp upturn in the resistivity associated with the Néel transition shows up as the superconductivity is suppressed with the magnetic field.

lish the doping-temperature phase diagram in Fig. 2. A peculiarity of $R\mathrm{Ba_2Cu_3O_{6+x}}$ crystals is that their doping level is determined both by the oxygen content and by the degree of its ordering [5, 6]. For characterizing the doping level, we use therefore the in-plane conductivity $\sigma_{ab}(280K)$ instead of the oxygen content; the former is a good measure of the hole density given that the hole mobility stays almost constant in the doping region under discussion [18].

As can be seen in Fig. 2, the long-range AF order in $R\mathrm{Ba_2Cu_3O_{6+x}}$ appears to be much more stable than in $\mathrm{La_{2-x}Sr_xCuO_4}$ where it vanishes well in advance before the onset of superconductivity. In $R\mathrm{Ba_2Cu_3O_{6+x}}$, the AF phase boundary flattens upon approaching the SC compositions and hits the SC region, crossing the T_c line at ≈ 15 K. The observed overlap of the SC and AF regions is very close to the area where μSR studies of $\mathrm{YBa_2Cu_3O_{6+x}}$ ceramics revealed the coexistence of superconductivity with spontaneous static magnetism [4, 7]. Given that the c-axis resistivity studied here is sensitive to the *interlayer* spin ordering, the static magnetism detected by μSR [4, 7] should in fact be related to the three-dimensional AF order. The superconductivity in $R\mathrm{Ba_2Cu_3O_{6+x}}$ thus develops directly from the AF-ordered state without any intervening paramagnetic or spin-glass region.

According to the established phase diagram (Fig. 2), an increase of the hole density in $\mathrm{CuO_2}$ planes by $\sim$ 1% per Cu (from $\approx 5\%$ to $\approx 6\%$, assuming the onset of superconductivity at $\approx 5\%$ doping) turns an AF $R\mathrm{Ba_2Cu_3O_{6+x}}$ crystal without any sign of superconductivity into a bulk superconductor with $T_c \sim 15$ K. What happens with the AF order upon entering the SC region, does it vanish abruptly? Zero-field $\rho_c(T)$ curves measured on the same $\mathrm{LuBa_2Cu_3O_{6+x}}$ crystal for two hole doping levels (that differ by $\approx$ 0.5-0.6% per Cu) indeed demonstrate a switch from an AF state with $\rho_c(T)$ sharply growing below T_N to a SC state (Fig. 3). However, when the superconductivity is suppressed with the magnetic field $\mathbf{H} \parallel \mathbf{c}$, the steep increase in ρ_c associated with the AF ordering is recovered back (Fig. 3). Moreover, the recovered Néel temperature is merely several Kelvin lower than for the non-SC composition (upper curve in Fig. 3) and the resistivity increase is not reduced appreciably either. As long as the SC in $\mathrm{LuBa_2Cu_3O_{6+x}}$ and $\mathrm{YBa_2Cu_3O_{6+x}}$ crystals is weak enough to be killed by the 16.5-T field, the unveiled $\rho_c(T)$ curves keep demonstrating the anomalous growth below 15-20 K asso-

ciated with the Néel transition. This behavior indicates that, at least when superconductivity is suppressed with magnetic fields, the AF order extends to considerably higher doping levels than the SC onset. Consequently, at zero magnetic field the AF and SC orders either coexist with each other in a certain range of doping, or the AF order is frustrated in the SC state but revives as the superconductivity is destroyed with the magnetic field. A switching between the AF and SC orders was indeed suggested based on early μSR studies [4], yet no further proofs were collected. The close location and even overlapping of the AF and SC orders on the phase diagram raise another question of whether the AF and SC orders reside in nanoscopically separated phases in CuO_2 planes or coexist on the unit-cell scale, which calls for local microscopic tools to be clarified.

Acknowledgments

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